

X-ray-induced atomic motion in phase-change materials and conventional covalent chalcogenide glasses

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X-ray photon correlation spectroscopy (XPCS) enables direct access to atomic-scale dynamics in disordered materials, revealing both spontaneous and x-ray-induced relaxation processes. Here, we study two compositionally similar alloy glasses near their glass transition temperatures: the phase-change material (PCM) $\text{Ge}_{15}\text{Sb}_{85}$ and the non-PCM alloy $\text{Ge}_{15}\text{Te}_{85}$. Both exhibit an x-ray-induced atomic motion, yet with markedly different responses. $\text{Ge}_{15}\text{Sb}_{85}$ undergoes an immediate transition to an x-ray-induced yielding state, characterized by stationary dynamics governed solely by the absorbed dose. In contrast, $\text{Ge}_{15}\text{Te}_{85}$ shows a progressive slowing down of the x-ray-induced dynamics, accompanied by a crossover from compressed to stretched exponential decay in the density autocorrelation functions. This behavior is consistent with the emergence of liquidlike collective motion as supported by de Gennes narrowing in the wave-vector dependence of the dynamics at length scales comparable with the first sharp diffraction peak. Unlike $\text{Ge}_{15}\text{Sb}_{85}$, this alloy does not reach a stationary regime within experimental timescales, implying that the yielding transition occurs only after thousands of seconds with the available dose rate. Its response is also temperature-dependent: at lower temperatures, the dynamics reflects intrinsic stress relaxation processes, whereas at higher temperatures, it becomes dose-controlled. These findings demonstrate that the dynamical response to x-ray excitation is not determined solely by chemical composition or bonding character, but results from the interplay between irradiation effects and internal stress relaxation pathways.

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I. INTRODUCTION

X rays are a fundamental tool for studying the intrinsic properties of a material [1]. Over the past decades, improvements in the brilliance and coherence of x-ray beams delivered in modern synchrotron sources have led to the development of powerful techniques to investigate the dynamical properties of matter, one of the most significant being x-ray photon correlation spectroscopy (XPCS) [2]. This method provides information on the collective microscopic rearrangements that occur in disordered materials at nanometric and atomic scales [2]. In XPCS, the dynamics is investigated by analyzing the

temporal fluctuations in the intensity of speckles, which are interference patterns generated by coherent x rays scattered from the material. By recording a series of speckle patterns, XPCS captures these fluctuations, offering a direct measure of the particle dynamics. Studies on the atomic motion in hard materials have shown that the response of these systems to the high fluxes of x-ray beams employed in XPCS can vary significantly depending on the material family [3–17].

While in crystalline [3] and glassy [4–9] alloys XPCS provides information on the atomic diffusion and collective motion in the material, unexpected results have been reported in more conventional glasses such as vitreous silica and germania [10]. In these materials, structural rearrangements are frozen in the glassy state, and the intrinsic dynamics would be too slow to be directly accessed during an experimental campaign. Still, some dynamics can be triggered by the x rays due to *radiolysis* [10]. In this case, the probed dynamics is induced by the interaction between the glass and the x rays, leading to a characteristic decorrelation time τ which decreases as the beam intensity increases. This process occurs when an x-ray photon is absorbed, creating an electron-hole pair that leads to atomic displacement and the formation of

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a long-lived exciton. The latter gives rise to bond breaking and further atomic displacement. Subsequent studies have expanded this investigation to a variety of materials, including borate glasses [11–13,16], sodium silicates [16], lithium borates [12], and chalcogenide glasses [14,17]. In particular, the studies on the oxide glass LiBO_3 [18] and the chalcogenide glass GeSe_3 [17] show that after a certain absorbed dose, the system reaches a stationary state of yielding, which is photoinduced by the x rays. The specific absorbed dose depends on the physicochemical characteristics of the material. Altogether, these previous works have shown that the way the x-ray-matter interaction occurs depends on the specific details of the material and can provide information on the network topology of the glass. After the perturbation, the glass relaxes in a way which depends on the nature of the system. In this context, the probed relaxation time, τ , still quantifies the characteristic timescale of the observed motion, but it refers to *beam-induced dynamics*.

In general, these studies collectively demonstrate that x rays can be employed not only to probe spontaneous electron density fluctuations but also to drive them, granting access to material properties that would otherwise be inaccessible using conventional techniques. The response of a glass to x-ray irradiation contains information on the structure and the microscopic relaxation processes active in the material [10]. Hence, x rays have emerged as a powerful probe to study spontaneous and induced dynamical fluctuations in complex systems in response to other physical parameters, such as temperature and pressure [17].

Despite extensive studies on metallic and oxide glasses, no XPCS studies have yet addressed glassy phase-change materials (PCMs), nor directly compared them with compositionally similar non-PCMs. Such a comparison is essential to unravel the respective roles of bonding and composition in the beam-induced response. PCMs (such as $\text{Ge}_{15}\text{Sb}_{85}$, $\text{Ge}_2\text{Sb}_7\text{Te}_5$, GeTe , and $\text{Ag-In-doped Sb}_2\text{Te}$) are functional alloys widely used in nonvolatile memory and photonic devices [19,20]. They can be rapidly and reversibly switched between amorphous and crystalline phases by Joule heating or optical pulses. Their strong property contrast between the two phases can be used for encoding data or manipulating optical beams for functional devices. Although PCMs share a covalent glassy nature with non-PCM chalcogenide alloys (e.g., $\text{Ge}_{15}\text{Te}_{85}$, GeSe , and GeSe_2), they differ remarkably in physical properties such as liquid fragility, structural relaxations, crystallization kinetics, and glass-forming abilities [19,20]. Recent work by some of the present authors revealed that PCMs, including $\text{Ge}_{15}\text{Sb}_{85}$, show a pronounced secondary β relaxation in the glassy state, faster than the main α -relaxation process, associated with loosely bonded regions and enhanced atomic mobility [21]. By contrast, when Sb is replaced by Te, the similar composition $\text{Ge}_{15}\text{Te}_{85}$ non-PCM shows virtually no β relaxation, implying a rigid bonding network without local fast dynamics. The striking difference in relaxation behaviors is demonstrated to correlate with their markedly different glass-forming abilities, thermal stabilities, and crystallization kinetics [21]. While $\text{Ge}_{15}\text{Te}_{85}$ is a good glass former with a relatively stable supercooled liquid region, $\text{Ge}_{15}\text{Sb}_{85}$ can easily crystallize on approaching the glass transition temperature.

In this work, we present the first direct comparison of the evolution of the collective atomic motion induced by intense x-ray radiation between the phase-change material $\text{Ge}_{15}\text{Sb}_{85}$ and the compositionally similar non-PCM $\text{Ge}_{15}\text{Te}_{85}$, investigated in the vicinity of their glass transition temperature T_g . $\text{Ge}_{15}\text{Sb}_{85}$ is a metallic-like alloy classified as a PCM due to its rapid and reversible glass-crystal switching properties, which are fundamental for nonvolatile memory applications [19,22,23]. $\text{Ge}_{15}\text{Te}_{85}$, on the other hand, is a conventional chalcogenide glass with high amorphous stability and slow crystallization kinetics, extensively studied in both liquid and glassy phases [22,24–32]. The strong interest in this composition is driven by its favorable properties for selector devices, such as a high crystallization temperature that ensures a wider operating range, a fast threshold switching speed enabling rapid off-to-on transitions, and a low threshold voltage for efficient switching [28]. This unique comparison demonstrates that the material's dynamical response to intense x-ray fluxes is not governed solely by chemical composition or bonding type, but emerges from the interplay between atomic mass, network topology, intrinsic relaxation channels, and absorbed dose.

Finally, XPCS offers a view that is complementary to other techniques commonly used to study phase-change materials. Ultrafast optical and time-resolved diffraction experiments [33–36] capture the temporal evolution of the structure during laser- or current-induced switching, revealing nonthermal bond breaking and transient crystalline order on femtosecond timescales. Neutron scattering, instead, accesses fast atomic diffusion in the liquid phase but remains insensitive to the slow, collective motion frozen in the glass. In contrast, XPCS provides a direct measurement of atomic dynamics—not of structural changes—by tracking temporal correlations in the scattered intensity. This makes it uniquely suited to probe the beam-induced motion within the amorphous state and to disentangle intrinsic relaxation processes from irradiation effects. The resulting beam-induced dynamics encode the material's intrinsic relaxation landscape, offering a powerful and complementary route to explore the atomic motion of glassy PCMs.

II. EXPERIMENTAL METHODS

A. Samples

Glassy samples of $\text{Ge}_{15}\text{Sb}_{85}$ and $\text{Ge}_{15}\text{Te}_{85}$ were prepared through magnetron sputtering deposition of stoichiometric targets synthesized following the same procedure described in a previous study [22]. A thick uniform layer of samples was deposited onto a 4-inch Si (100) wafer using DC 10 W, 10 sccm Ar flow, and 5×10^{-3} mbar Ar pressure. A steep-edge profilometer determined the sputtering rates of the target compositions to estimate the sputtering time needed for the desired layer thickness L (3–8 micrometers) to match the absorption length μ of the samples. The amorphous layers were separated from the Si substrate in the form of pieces or flakes, where single relatively large pieces (with a diameter of several millimeters) were selected for XPCS measurements. The nominal sample compositions were confirmed (within instrumental error $\sim 3\%$) twice using x-ray fluorescence (XRF)

TABLE I. Calorimetric glass transition temperature T_g , wave vector of the first sharp diffraction peak (FSDP) maximum, mass density ρ , sample thickness, and x-ray absorption length at the energies used in the XPCS experiments for $\text{Ge}_{15}\text{Te}_{85}$ and $\text{Ge}_{15}\text{Sb}_{85}$ [19,21,27].

	T_g (K)	FSDP (nm^{-1})	ρ (g/cm^3)	L (μm)	μ (μm)
$\text{Ge}_{15}\text{Sb}_{85}$	500	20.5	5.323	3	8.5
$\text{Ge}_{15}\text{Te}_{85}$	412	19.6	5.53	8	6.9

with a Rigaku NEX CG, first for the sputtering target and then for the as-deposited layers. The main properties of the two samples are summarized in Table I [19,21,27].

Although our study has been performed on thin layers, we expect that the underlying physics governing relaxation and photoinduced dynamics remains the same, since the glassy state, regardless of the preparation route, is always a metastable configuration, being the glass transition a kinetic process. Consequently, only the absolute values of the relevant quantities (such as relaxation times) may vary, while the fundamental mechanisms behind the beam-induced dynamics remain unchanged.

B. X-ray photon correlation spectroscopy

XPCS experiments were carried out during two separate beamtimes at the beamline P10 of PETRA III synchrotron in Hamburg, Germany. In the first experiment, we measured the $\text{Ge}_{15}\text{Te}_{85}$ composition. An x-ray beam of 8 keV was focused onto a spot with a full width at half maximum (FWHM) of $2.8 \times 2.2 \mu\text{m}^2$ ($H \times V$) with a photon flux of $\phi_0 \sim 4.5 \times 10^{10}$ ph/s, whereas in the second experiment, we measured the $\text{Ge}_{15}\text{Sb}_{85}$ using a beam energy of 8.1 keV, with a spot size of $3.4 \times 2.0 \mu\text{m}^2$ and a photon flux of $\phi_0 \sim 8 \times 10^{10}$ ph/s. The samples were placed under vacuum to prevent oxidation on a Cu sample holder, whose temperature was controlled by a resistive heater and a Lakeshore temperature controller. Measurements were performed at a wave vector q corresponding to the position of the maximum of the FSDP for both compositions. $\text{Ge}_{15}\text{Te}_{85}$ was studied at temperatures $T = 295$ and 325 K (corresponding to $T \simeq [0.7, 0.8]T_g$, see values in Table I), while $\text{Ge}_{15}\text{Sb}_{85}$ was measured at $T = 295, 350$, and 400 K ($[0.6, 0.7, 0.8]T_g$). Additionally, the $\text{Ge}_{15}\text{Te}_{85}$ composition was investigated as a function of the scattering vector q in the range $[2.18\text{--}27.37] \text{ nm}^{-1}$ at 295 K.

To explore the effect of the x rays on the dynamics, the full beam intensity ϕ_0 was attenuated through Si attenuators (att). The fractions of the transmitted beam intensity (i.e., the transmission T_a) and the relative beam fluxes ϕ used in the experiments are summarized in Table II. It is important to note that the x-ray beam can also induce a slight local heating of the sample, typically limited to a few degrees, as also measured by Baglioni *et al.* for GeSe_3 by coupling XPCS with fast differential scanning calorimetry [17]. Although a direct measurement of this local temperature rise is not accessible under our experimental conditions, we can reasonably consider a similar thermal gradient in our sample as we have employed the same experimental configuration as in Ref. [17].

TABLE II. Transmission percentages for the silicon attenuators used in the XPCS experiments for each material.

$\text{Ge}_{15}\text{Sb}_{85}$			$\text{Ge}_{15}\text{Te}_{85}$		
	T_a	ϕ ($\times 10^{10}$ ph/s)		T_a	ϕ ($\times 10^{10}$ ph/s)
att = 0	100%	8	att = 0	100%	4.5
att = 2	45%	3.58	att = 2	44%	1.96
att = 4	22%	1.73	att = 3	28%	1.26
att = 6	10%	0.77	att = 4	20%	0.91
att = 8	5%	0.40	att = 6	9%	0.39

Each XPCS measurement was performed on a fresh sample spot to avoid any cumulative radiation effects. Different series of speckle patterns were collected in transmission geometry using an EIGER 4M detector positioned 1.8 m from the sample stage. Fluctuating speckle patterns are obtained when an amorphous system scatters a coherent x-ray, and they reflect the internal dynamics of the sample. Thus, the autocorrelation of the scattered intensity allows for the calculation of the microscopic dynamics, being

$$C_q(t_1, t_2) = \frac{\langle I_p(t_1)I_p(t_2) \rangle_p}{\langle I_p(t_1) \rangle_p \langle I_p(t_2) \rangle_p}. \quad (1)$$

This function is called *two-time correlation function* (TTCF). $I_p(t)$ is the intensity measured at pixel p and time t , and $\langle \dots \rangle_p$ denotes an average over detector pixels at a given momentum transfer q . Examples of TTCFs are presented in Figs. 2(c)–2(d). The main diagonal of the TTCFs corresponds to $t_1 = t_2$ and represents the correlation of a frame at a given time with itself, while all the off-diagonals are the correlation of a frame at time t_1 (or t_2) with a subsequent frame at time t_2 (or t_1).

From $C_q(t_1, t_2)$, the autocorrelation function $g_2(t)$ of the intensity fluctuations can be extracted:

$$g_2(q, t) = \langle C_q(t_0, t_0 + t) \rangle, \quad (2)$$

where $t_0 = t_1$, $t = t_2 - t_1$, and $\langle \dots \rangle$ represents the average over time. This means that the TTCFs are a map of the time evolution of the microscopic dynamics. The $g_2(q, t)$ provides information on the microscopic dynamics through the determination of the *intermediate scattering function* (ISF) $F(q, t)$ via the Siegert relation $g_2(q, t) = 1 + C_e |F(q, t)|^2$, where C_e is the experimental contrast related to the degree of coherence of the x rays [37]. The ISF is the time-dependent density-density correlation function and quantifies the extent to which the atomic configuration at a given moment resembles its configuration at a later time t .

III. RESULTS

We model the $g_2(q, t)$ with a Kohlrausch-Williams-Watts (KWW) function [38]

$$g_2(q, t) = y_0 + C e^{-2\left(\frac{t}{\tau}\right)^\beta}, \quad (3)$$

where $C = C_e |f_q|^2$, with f_q being the non-ergodicity factor and C_e the experimental contrast, τ is the relaxation time, β the degree of exponentiality of the decay, and y_0 accounts for the baseline [37]. All parameters depend on the exchanged wave vector q , the temperature T , and the absorbed dose D .

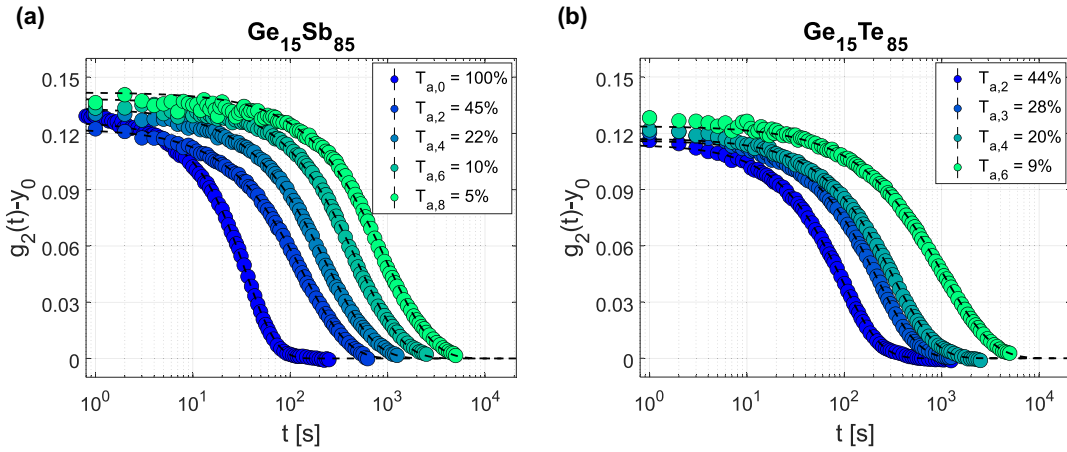


FIG. 1. Auto-correlation functions of the intensity fluctuations measured with XPCS at $0.7T_g$ in (a) $\text{Ge}_{15}\text{Sb}_{85}$ and (b) $\text{Ge}_{15}\text{Te}_{85}$ at varying beam attenuations. The curves of each sample are obtained by ensuring a total absorbed dose of 38 GGy for both samples. The dashed black lines represent the best fits to the KWW model. Each curve has been measured on a fresh sample position.

The total radiation dose absorbed by the sample during an exposure time Δt is calculated as

$$D = \frac{\epsilon}{m} F \Delta t, \quad (4)$$

where ϵ is the x-ray energy and F is the fraction of the incident beam, ϕ , absorbed by the sample given by $F = \phi(1 - T_r)$, with T_r being the sample transmission. As anticipated in the experimental section, the value of the incident flux ϕ was varied by attenuating the full beam ϕ_0 through Si attenuators with transmissions T_a reported in Table II. The incident fluxes employed in the experiments result in $\phi = \phi_0 T_a$. The dose also depends on the mass m within the scattering volume V_s :

$m = \rho V_s$, where $V_s = AL$ (with A as the focal spot area and L the sample thickness) and ρ is the mass density.

Consistent with previous studies on other chalcogenide glasses [14,17], our data show that the probed atomic motion is also triggered by the x-ray irradiation in the studied compositions. This effect is observed at all investigated temperatures and at the probed q , no appreciable structural damage is detected (see Fig. S1 in Supplemental Material [39]). However, within the limits of our experimental resolution, we cannot determine whether this apparent stability reflects a genuine resistance to structural damage, as reported for other glassy systems [10], or whether minor structural damage occurs below our detection threshold or in a different q range.

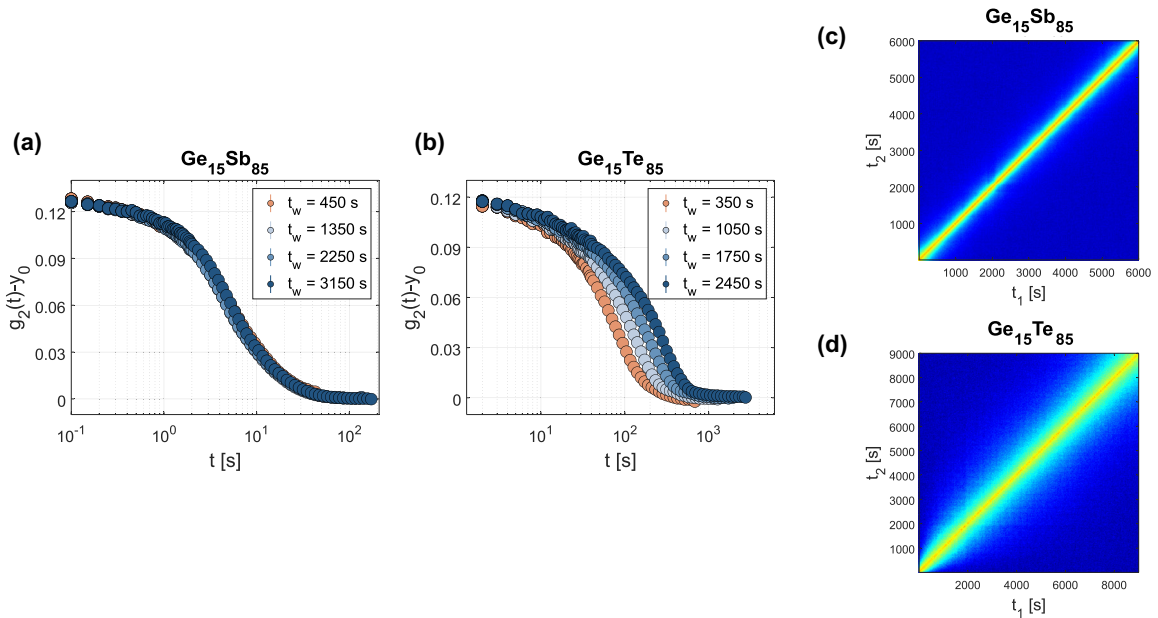


FIG. 2. Temporal evolution of the dynamics at $0.7T_g$ for a fixed flux of the x-ray beam of 3.58×10^{10} ph/s in $\text{Ge}_{15}\text{Sb}_{85}$ (a) and of 1.96×10^{10} ph/s in $\text{Ge}_{15}\text{Te}_{85}$ (b). The dynamics remains stationary within the probed time window for $\text{Ge}_{15}\text{Sb}_{85}$ (a), while physical aging in $\text{Ge}_{15}\text{Te}_{85}$ is signaled by the shift of the curves toward longer times with increasing t_w (b). Two-times correlation functions measured in $\text{Ge}_{15}\text{Sb}_{85}$ (c) and $\text{Ge}_{15}\text{Te}_{85}$ (d) showing stable dynamics and aging, respectively.

An example of the probed dynamics is shown in Fig. 1 where we report autocorrelation functions measured with XPCS at different values of the x-ray flux at $T = 0.7T_g$, for both $\text{Ge}_{15}\text{Sb}_{85}$ [Fig. 1(a)] and $\text{Ge}_{15}\text{Te}_{85}$ [Fig. 1(b)]. The long-time decay of the curves corresponds to the characteristic time associated with the probed dynamics. In both cases, this decay shifts to larger times with decreasing flux (from blue to green curves), indicating that the dynamics can be tuned by varying the intensity of the x-ray beam. The black dashed lines are the best fits to the experimental data according to Eq. (3). To ensure consistency between data collected at different x-ray fluxes, the comparison of the curves was performed by evaluating the $g_2(t)$ considering a fixed absorbed dose of 38 GGy for each composition. Although the dynamics of both glasses clearly depends on the x-ray flux, the two materials exhibit distinct responses to x-ray exposure. This is particularly evident from the temporal evolution of their relaxation processes at a fixed beam intensity as shown in Fig. 2. Here, the curves were evaluated by averaging the TTCFs according to Eq. (2) over time windows Δt , centered at waiting time $t_w = \Delta t(n + 1/2)$ with $n \in [0, 3]$, and $\Delta t = 900$ s for the $\text{Ge}_{15}\text{Sb}_{85}$ and $\Delta t = 700$ s for the $\text{Ge}_{15}\text{Te}_{85}$. In this context, the waiting time t_w reflects the time elapsed since the onset of each measurement. As depicted in Fig. 2(a), $g_2(t)$ curves acquired at different waiting times overlap each other, indicating that the dynamics of the $\text{Ge}_{15}\text{Sb}_{85}$ remains stationary over time. This is also evident from the TTCF, shown in Fig. 2(c), where a constant intensity profile is observed along the main diagonal from the bottom left corner to the upper right corner. In contrast, $\text{Ge}_{15}\text{Te}_{85}$ exhibits a progressive slowing down of its dynamics over time. As shown in Fig. 2(b), the $g_2(t)$ curves shift toward longer decay times as the waiting time progresses from 350 s to 2450 s, highlighting this effect. The presence of this process is also confirmed by the broadening of the intensity along the main diagonal of the TTCF of $\text{Ge}_{15}\text{Te}_{85}$ reported in Fig. 2(d).

The slowing down of the dynamics is a spontaneous process in glasses held at temperatures close to T_g , known as physical aging [40]. As out-of-equilibrium systems, glasses naturally evolve over time toward more energetically stable configurations, leading to an increase in relaxation time. Under standard conditions, this aging process is thermally driven, as the system progressively explores its energy landscape. In our experiments, however, the observed slowing down is not solely spontaneous but is induced by the x-ray beam itself. The beam not only probes and stimulates atomic dynamics but also promotes a temporal evolution of the system. This effect may be linked to stress release occurring under irradiation or to local annealing processes initiated by the beam. These aspects will be discussed in greater detail later in this section.

The contrasting temporal evolutions of the dynamics in the two compositions imply a different response of the two systems to the accumulated dose D . In order to compare measurements performed under different beam fluxes, or on the two different samples, it is important to take into account all the parameters influencing the dynamics. For this reason, τ alone is not the most effective parameter to consider, instead, it is more meaningful to normalize τ to the absorbed photon flux, photon energy, and mass within the scattering volume. For this purpose, we express the relaxation time τ , obtained

from XPCS measurements and KWW fits to Eq. (3), in terms of the absorbed dose D_τ required to induce the observed dynamics:

$$D_\tau = \frac{\epsilon}{m} \phi_0 T_a (1 - T_r) \tau. \quad (5)$$

In this way, D_τ represents the absorbed dose during one relaxation time under constant beam flux and transmission, which is indeed the case for the measurements presented here. This formulation accounts for the interaction between the material and the x rays and it provides an expression for τ that is independent of beam intensity, energy, or sample characteristics (see also Supplemental Material [39]). In the case of a process purely induced by irradiation [10], the relaxation time τ is expected to scale inversely with the incident x-ray flux ϕ , meaning that halving the beam flux ($T_a = 50\%$) doubles τ . Using this expression, we can compare the relaxation dynamics of the two probed compositions. To explore the distinct responses of $\text{Ge}_{15}\text{Sb}_{85}$ and $\text{Ge}_{15}\text{Te}_{85}$ to x-ray exposure, Figs. 3 and 4 show the absorbed dose in relaxation time D_τ and the KWW exponent β as functions of the accumulated dose D , for different attenuator transmissions $T_{a,i}$. This representation enables us to assess the dependence of the dynamical parameters on the absorbed dose and provides insight into the nature of the interaction, allowing us to distinguish between dynamics that are purely beam-induced and those influenced by additional mechanisms.

First, we focus on the dynamical behavior of $\text{Ge}_{15}\text{Sb}_{85}$. The dependence of the XPCS parameters τ and β on the accumulated dose D is presented in Fig. 3 at two temperatures, $0.7T_g$ or $0.8T_g$. Panels (a) and (c) show the dose D_τ accumulated in a relaxation time τ at the two temperatures, while the dose evolution of the β parameter is plotted in panels (b) and (d). Different symbols and colors refer to different beam fluxes. The probed dynamics is beam-induced, since the D_τ parameter is independent of the beam flux for almost all the datasets, implying that the decorrelation time τ is inversely proportional to the beam flux. Overall, a good collapse of points corresponding to different beam fluxes is observed in all panels. There is, however, one dataset that follows a different trend. The blue circles at $T = 0.7T_g$, corresponding to full beam on the sample, exhibit a lower D_τ and a higher β compared to the other points, likely due to local beam-induced heating that slightly increases the effective temperature of the probed spot. Apart from this dataset, the other points suggest that the system is in a yielded state at all the investigated doses, because the D_τ parameter is almost constant during the measurement and the stretching parameter β is close to one, as in the case of LiBO_3 [18] and GeSe_3 [17]. We now turn to the dynamical properties of $\text{Ge}_{15}\text{Te}_{85}$. As discussed before, a purely beam-induced dynamics is expected to scale with the accumulated absorbed dose D : if the dynamics is entirely governed by the x rays, parameters such as the absorbed dose in XPCS relaxation times, D_τ , and the shape parameter β should collapse onto universal curves when plotted as a function of D . However, for $\text{Ge}_{15}\text{Te}_{85}$ at $0.7T_g$, this universal scaling is not observed (see Fig. S2). Although the beam induces the dynamics, D_τ and β exhibit significant variations with the incident flux when plotted against D , suggesting that

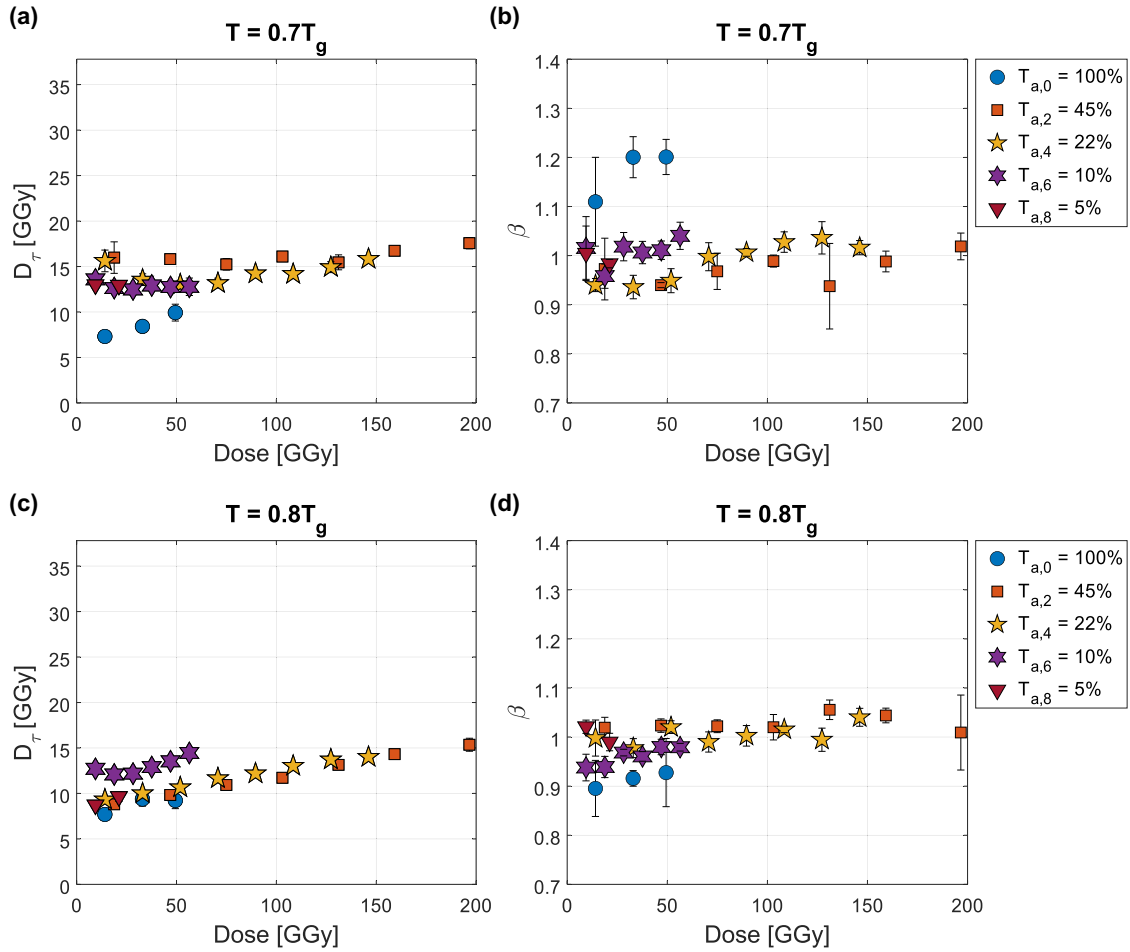


FIG. 3. Evolution of the dynamics in $\text{Ge}_{15}\text{Sb}_{85}$ as a function of the absorbed dose. [(a) and (b)] At $0.7T_g$ and [(c) and (d)] at $0.8T_g$, the D_τ and the β tend to collapse into a master curve when plotted versus dose. Both parameters are almost independent of the applied dose.

the absorbed dose alone does not fully describe the system's evolution.

To further clarify this behavior, we plot D_τ and β as functions of the experimental waiting time t_w , defined as the time elapsed since the start of exposure. Interestingly, when expressed in terms of t_w , the data collapse onto master curves across all fluxes [Figs. 4(a) and 4(b)], revealing a universal, time-dependent evolution of the dynamics. To our knowledge, such a collapse with respect to the waiting time, rather than the absorbed dose, has not been previously reported in studies of beam-induced dynamics. This observation indicates that, while x-ray irradiation initiates atomic motion, the evolution at $0.7T_g$ remains governed primarily by intrinsic relaxation processes with timescales dictated by the material itself, rather than solely by the accumulated dose.

It is important to note that the slowing down of the dynamics is not attributed to a conventional structural relaxation, but rather to a local yielding transition induced by the x-ray beam. The irradiation creates localized stress sources, similar to Eselby's inclusions, that lead to a displacement field, giving rise to the observed dynamics. At higher absorbed doses, the density of these inclusions becomes sufficiently large to trigger a yielding transition and the onset of liquidlike behavior. This process has been well verified in oxide glasses, where the x-ray beam drives the sample into a new configuration, similar

to a rejuvenated, high enthalpy, glass [18]. At high absorbed doses, the x-ray beam fluidizes the glass, which behaves as a supercooled liquid. The corresponding increase in D_τ can thus be interpreted as a relaxation toward a new, high-enthalpy glass configuration, and the structural relaxation process is completely hindered by the induced dynamics, given that $\tau_\alpha \gg \tau_{\text{induced}}$ at the probed temperatures.

Further evidence supporting this interpretation comes from “beam-off” experiments (Fig. 4 insets), where the x-ray beam was switched off for extended periods (see Fig. S3 in Supplemental Material [39] for further details). For short interruptions (10 minutes), stress relaxation continues. In contrast, for longer interruptions (1 hour), both D_τ and β reverted toward their initial values, indicating a recovery of the original state in the absence of irradiation. Such a reversible response upon switching off the beam has never been observed in stationary beam-induced dynamics so far, and supports the absence of structural damage in the dose regimes here probed.

In contrast to previous observations (e.g., Martinelli *et al.* [18]), where such transitions were accompanied by permanent structural changes, our measurements in $\text{Ge}_{15}\text{Te}_{85}$ show no detectable variation in scattered intensity throughout the irradiation, indicating that the magnitude of the locally generated stresses is insufficient to produce measurable structural modifications. This suggests that the transition is incomplete

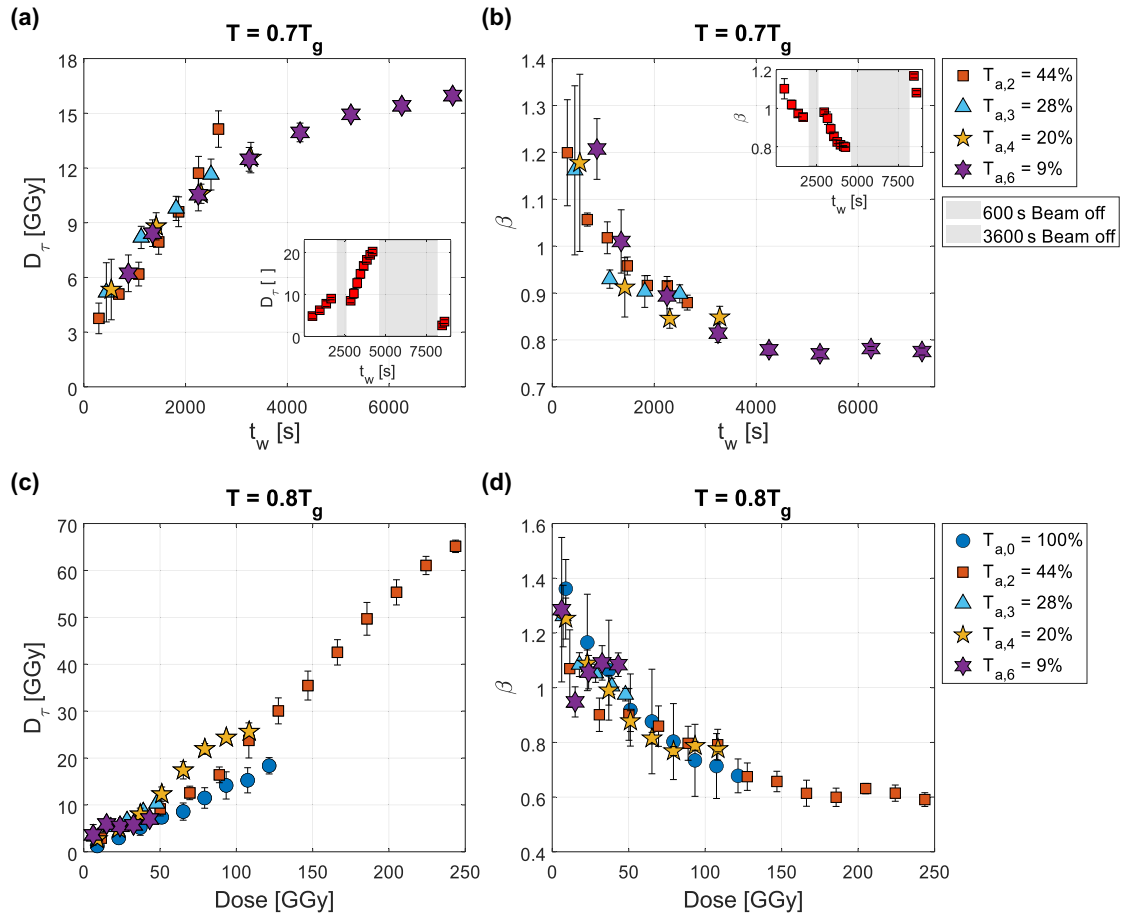


FIG. 4. [(a) and (b)] Evolution of the dynamics in $\text{Ge}_{15}\text{Te}_{85}$ as a function of the waiting time t_w , at $0.7T_g$. D_τ and β collapse into a master curve when plotted vs t_w , suggesting the dynamics is primarily influenced by intrinsic relaxations active in the material. While the beam does not irradiate the sample spot, both D_τ and β tend to recover the initial state [insets of (a) and (b)]. [(c) and (d)] At $0.8T_g$, D_τ and β collapse onto a master curve when plotted versus dose. [(a) and (c)] As the waiting time or the deposited dose increases, the dynamics slows down, suggesting a final plateau. [(b) and (d)] With increasing absorbed dose, the shape parameter β decreases from $\beta > 1$, a signature of compressed correlation functions, to $\beta < 1$, typical of stretched exponential decays.

and that the induced stresses are below the threshold required for a permanent rearrangement. Consequently, the effect is partially reversible: once the beam is switched off, the material gradually releases the accumulated internal stresses over a characteristic timescale. The response is thus not purely elastic, and the recovery of the initial dynamical state requires a finite time.

The hypothesis of room-temperature stress release is confirmed by the fact that at $0.8T_g$, both D_τ and β exhibit a universal scaling with the absorbed dose D , regardless of the incident flux [Figs. 4(c) and 4(d)]. This indicates that, at $0.8T_g$, the dynamics depends solely on the x-ray flux and is thus *purely* beam-induced. Remarkably, a modest 10% increase in temperature is sufficient to eliminate the stress-driven contributions observed at $0.7T_g$, placing the system entirely under the control of irradiation-driven dynamics.

We thus have evidence of the coexistence of two mechanisms. On the one hand, an x-ray-induced process of rejuvenation appears as the system approaches the onset of the yielding transition, which occurs at both investigated temperatures. On the other hand, at $0.7T_g$, an additional stress release related to sample preparation, possibly triggered by a slight

local heating of a few degrees, takes place. This secondary contribution affects the scaling behavior when plotting D_τ or β versus D , and makes the behavior at $0.7T_g$ somewhat different from that observed at $0.8T_g$. At room temperature ($0.7T_g$) the x-ray-induced tendency toward a yielding transition coexists with a sample-specific stress-relaxation process, and the latter modifies the scaling behavior. At $0.8T_g$, where this residual stress release is absent thanks to a 10% heating, the dynamics is governed solely by the beam and the D_τ versus D scaling is recovered.

Next, we examine how the dynamical parameters of the two compositions depend on the absorbed dose D (or on the waiting time t_w) at both $0.7T_g$ and $0.8T_g$. For this purpose, we refer to Figs. 3 and 4 for $\text{Ge}_{15}\text{Sb}_{85}$ and $\text{Ge}_{15}\text{Te}_{85}$, respectively. As anticipated, the $\text{Ge}_{15}\text{Sb}_{85}$ glass exhibits stationary dynamics and a KWW exponent $\beta \lesssim 1$ at both investigated temperatures. These features correspond to a local state of photoinduced yielding by the x rays, resembling the previous studies on other chalcogenide and oxide glasses [14,17,18]. In materials science, yielding refers to the transition from an elastic response, where deformation is reversible, to a plastic state, where the material undergoes irreversible atomic

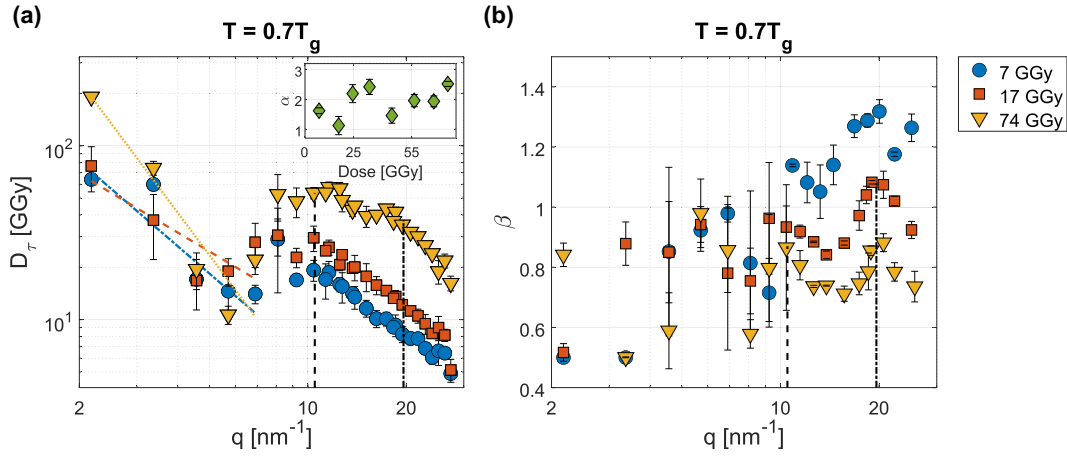


FIG. 5. Wave vector dependence of τ (a) and β (b) at $0.7T_g$ of $\text{Ge}_{15}\text{Te}_{85}$ for three selected doses: $D = 7$ GGy (blue circles), $D = 17$ GGy (red squares), and $D = 74$ GGy (yellow triangles). The vertical dashed and dashed-dotted black lines represent, respectively, the position of the prepeak and the main peak. (a) The inset shows the dose dependence of the power-law exponent α describing the linear dependence of τ at low q values.

rearrangements. In glasses, yielding is particularly subtle since plastic events appear at very small scales and are associated with localized atomic rearrangements known as shear transformation zones [18]. Martinelli *et al.* [18] showed that x-ray irradiation acts like an external force, generating tiny defects that trigger these hidden structural rearrangements, effectively pushing the material into a plastic, irreversible state at the microscopic scale. In the case of the chalcogenide glass GeSe_3 , the yielding is induced within 100 s of exposure at ambient temperature [17], while the $\text{Ge}_{15}\text{Sb}_{85}$ composition reaches this stationary state within the exposure time of 1 s at the probed temperatures of 350 K and 400 K ($0.7T_g$ and $0.8T_g$, respectively). This difference in timescales is likely the consequence of the larger thermal fluctuations at the probed temperatures, which facilitate the occurrence of local yielding at a lower dose. In fact, in agreement with previous works, we also observe a transitory state with a weak aging at ambient temperature ($0.6T_g$) in $\text{Ge}_{15}\text{Sb}_{85}$ accompanied by a transition in the KWW parameter β from compressed (≥ 1) to stretched (≤ 1) and reaching a stationary regime around 40 GGy (see Fig. S4). As discussed in detail in Ref. [18], these are the typical characteristics of the induced yielding transition.

On the other hand, the $\text{Ge}_{15}\text{Te}_{85}$ exhibits a strong dependence on the waiting time and absorbed dose at both temperatures, as evident from Fig. 4. We see that neither the absorbed dose associated with the relaxation time D_τ nor the β parameter reaches a complete plateau. Only the D_τ measured at $T_{a,6}$ and the β at $T_a \geq 28\%$ at $0.7T_g$ suggest the proximity to a stationary regime. To understand whether this behavior corresponds to the onset of the photoinduced yielding transition, we measured the wave-vector q dependence of the dynamics (and thus of the parameters D_τ and β) as a function of increasing absorbed dose (see Fig. 5). Following Ref. [18], during the transition to local yielding, one should also observe a crossover from a $1/q$ wave-vector dependence of τ and an almost q -independent evolution of β , to a liquidlike behavior. At low q , a linear trend emerges in the log-log plot of D_τ versus q [Fig. 5(a)], indicating a power-law relationship between

the relaxation time and the wave vector. By analyzing this dependence in the range $[2-7] \text{ nm}^{-1}$, we can access the nature of the particle dynamics and how it evolves with absorbed dose. Fitting the data with the expression $D_\tau(q) = (bq^\alpha)^{-1}$, where the exponent α characterizes the type of motion, we obtain $\alpha = 1.7 \pm 0.4$, independently of dose [see inset of Fig. 5(a)]. This value is larger than the one reported for LiBO_2 glass, where α decreases from ~ 1.1 to ~ 0.3 with increasing dose. This trend was interpreted as a transition in the nature of collective motion, from ballistic-like dynamics to a regime where defect creation and annihilation are balanced, similar to a steady-state condition observed in liquids [18]. Differently, in the $\text{Ge}_{15}\text{Te}_{85}$, the larger value of α suggests the presence of diffusive motion at low q values, with a negligible dose dependence. Additional data at lower q values, well separated from structural contributions, are required to confirm this behavior.

At high q (greater than 7 nm^{-1}), we observe an overall increase in D_τ and a concurrent decrease in β with increasing dose D . At all doses, D_τ presents a peak at $\sim 10.5 \text{ nm}^{-1}$, which mirrors the corresponding prepeak of the static structure factor $S(q)$ [27]. At high doses, a peak arises at $\sim 19.6 \text{ nm}^{-1}$, corresponding to the main first sharp diffraction peak (FSDP). Even the KWW β parameter exhibits features resembling the evolution of the $S(q)$, less evident at low doses but pronounced at higher doses. The resemblance between the q dependence of τ and β and that of $S(q)$ is reminiscent of the phenomenon known as *de Gennes narrowing*, which is named for the narrowing at q values corresponding to the FSDP of the width of the quasielastic contribution in neutron scattering experiments on liquids [41]. Given that the half-width at half maximum of the quasielastic peak is inversely proportional to structural relaxation time, $\Gamma(q) \propto 1/\tau$, this results in an increase in τ near structure factor peaks, implying that the most probable structural configuration is also the most stable. Therefore the observed q dependence in $\text{Ge}_{15}\text{Te}_{85}$ suggests that the sample volume irradiated by the x rays displays a liquidlike behavior and thus local yielding, as in LiBO_2 . This

interpretation is supported by the transition of β from values greater than 1, typical of a stress-driven dynamics of glasses well below T_g [8], to values lower than 1, a signature of liquidlike heterogeneous relaxation dynamics [6,42].

We note also that at low doses (below 0.5 GGy), the de Gennes narrowing is entirely absent for LiBO_2 , in both τ and β [18]. In our case, instead, the de Gennes narrowing effect is already present at the 10.5 nm^{-1} prepeak in τ (D_τ), even in the lowest probed dose regime (below 7 GGy). In particular, at low doses only the signature of the prepeak around $\sim 10.5 \text{ nm}^{-1}$ is observed in the evolution of D_τ with q , while at high doses, the signature of the main peak around $\sim 19.6 \text{ nm}^{-1}$ arises as well. Such a strong dynamical signature corresponding to the weak prepeaks in structure factors is a general feature of glass-forming liquids, which has been associated with the influence of dynamic heterogeneity phenomena at the single- and many-particle levels [43,44].

IV. DISCUSSION AND CONCLUSION

Our results reveal a strong correlation between the local atomic structure and the dynamical response to intense x-ray fluxes in two compositionally similar glasses, $\text{Ge}_{15}\text{Sb}_{85}$ and $\text{Ge}_{15}\text{Te}_{85}$. Consistent with previous findings in other oxides and chalcogenides [14,17,18], the x rays can induce a local state of yielding in both compositions, where the amorphous solids present liquidlike features in the irradiated zone. The propensity for this phenomenon, however, strongly depends on the chemical and physical details of the material.

In the PCM $\text{Ge}_{15}\text{Sb}_{85}$, our data exhibit an almost instantaneous photoinduced yielding transition at $0.7T_g$ and $0.8T_g$. This rapid response is favoured by thermal motion, since at room temperature the transition requires $\sim 400 \text{ s}$ of continuous irradiation to occur (see Supplemental Material [39]). This is in agreement with what has been observed in GeSe_3 [17] at ambient temperature, where yielding occurs in $\sim 100 \text{ s}$. The faster transition at room temperature in GeSe_3 is likely because Se is lighter than Sb, a fact supported by a faster relaxation time of $\tau \sim 2 \text{ s}$ in GeSe_3 in comparison to $\tau \sim 100 \text{ s}$ in $\text{Ge}_{15}\text{Sb}_{85}$ for similar doses. Increasing the temperature to 350 K ($0.7T_g$) enhances atomic mobility within $\text{Ge}_{15}\text{Sb}_{85}$, enabling the system to reach the yielding state almost immediately.

Unlike $\text{Ge}_{15}\text{Sb}_{85}$ and GeSe_3 , $\text{Ge}_{15}\text{Te}_{85}$ does not completely reach the yielding state within the experimental timeframe at neither $0.7T_g$ (which corresponds to room temperature) nor $0.8T_g$. This can be explained by the fact that Te atoms are heavier than Se or Sb atoms, reducing atomic mobility and resulting in a greater resistance to structural change upon irradiation with x-ray beams. This is consistent with the absence of visible structural changes in the FSDP, which are instead clear in GeSe_3 . Moreover, $\text{Ge}_{15}\text{Te}_{85}$ exhibits recovery effects after the beam is switched off: once the x-ray beam is turned off, the system begins to recover toward its initial dynamical state and the longer the absence of irradiation, the more closely the dynamical parameters return to their original values [see insets of Figs. 4(a) and 4(b)]. In contrast, the GeSe_3 , once it has reached the yielding state, shows stable dynamics: during the absence of the beam, the system remains in that state. It is important to emphasize that the onset and nature

of yielding under irradiation is likely determined by different factors, including the type of chemical bonding, the atomic mass, network topology, and local structural heterogeneity. Although both GeSe_3 and $\text{Ge}_{15}\text{Te}_{85}$ are covalent glasses with good glass-forming ability and slow crystallization kinetics, their microscopic bonding environments are different, with Ge-Te bonds weaker than Ge-Se bonds and having a lower bond energy [45]. Structural studies of amorphous GeSe_3 [46] have shown that its structural motifs are dominated by tetrahedra (the fraction of homopolar Ge-Ge or Se-Se bonds depending on preparation conditions). In contrast, amorphous $\text{Ge}_{15}\text{Te}_{85}$ contains both tetrahedral and defective-octahedral motifs, and the presence of Ge-Ge homopolar bonds has been reported [47]. Such structural and bonding distinctions are likely to contribute to the distinct physical behaviors observed between the two glass systems.

The faster photoinduced atomic motion in the PCM $\text{Ge}_{15}\text{Sb}_{85}$ and the greater sensitivity to x-ray-induced yielding are likely the consequence of its locally loosely bonded microscopic structure and the presence of a pronounced secondary β relaxation, which promotes local mobility at the atomic scale [21]. This mobility is also confirmed by the fact that the dynamics does not scale completely with the dose D , strengthening the idea of the presence of an intrinsic dynamic process already active at the probed temperatures. By contrast, the rigid covalently bonded network in $\text{Ge}_{15}\text{Te}_{85}$ is less prone to changes of the local atomic environment, which is reflected in longer relaxation times at the atomic scale and purely beam-induced dynamics but at ambient temperature. At this temperature, the scaling law is not fulfilled, probably due to the presence of some additional stresses related to the sample preparation, which combine with the irradiation-induced yielding transition.

Interestingly, we note that the $\text{Ge}_{15}\text{Te}_{85}$ exhibits a dependence on the absorbed dose similar to that reported in the oxide glass LiBO_2 [18]. In both systems, increasing the absorbed dose leads to a transition in the KWW β shape parameter from a compressed ($\beta \geq 1$) to a stretched ($\beta \leq 1$) regime, which is accompanied by the emergence of a liquidlike wave-vector dependence of the dynamics at large doses. As previously discussed, this behavior can be attributed to the transition from a purely beam-induced stress-driven motion to a local yielding transition of the material under x-ray irradiation [18].

It is, however, important to underline the main differences between LiBO_2 and $\text{Ge}_{15}\text{Te}_{85}$. One crucial factor influencing beam-induced dynamics is the x-ray absorption. $\text{Ge}_{15}\text{Te}_{85}$ consists of heavy atoms, which absorb x rays more efficiently than the lighter elements present in LiBO_2 . This difference is reflected in the experimental conditions: the thickness of the $\text{Ge}_{15}\text{Te}_{85}$ sample was comparable to its attenuation length at 8 keV ($\sim 8 \mu\text{m}$), ensuring substantial absorption, whereas LiBO_2 was significantly thinner ($125 \mu\text{m}$) relative to its attenuation length at 8.4 keV ($680 \mu\text{m}$). Consequently, the dose range explored for $\text{Ge}_{15}\text{Te}_{85}$ reached tens of GGy, while for LiBO_2 , it remained at a few GGy. Despite this large difference in the absorbed dose, both materials exhibit a similar evolution of the KWW β parameter and $\tau(q)$ as a function of the absorbed dose. This suggests that atomic rearrangements in $\text{Ge}_{15}\text{Te}_{85}$ require a significantly higher number of

electron-hole pairs, likely due to its heavier atomic composition. This interpretation aligns with the slower dynamics observed in $\text{Ge}_{15}\text{Te}_{85}$ (on the order of hundreds of seconds) compared to LiBO_2 (~ 20 s), despite its higher absorbed dose.

In conclusion, our study combines XPCS measurements at multiple temperatures with dose-resolved analysis, allowing a direct comparison between phase-change materials and covalent chalcogenide glasses. This approach enables us to uniquely probe the onset of x-ray-induced atomic motion in glasses and to distinguish it from sample-specific stress relaxation processes, which has not been addressed in earlier studies. Our work reveals distinct beam-induced dynamical responses in glassy $\text{Ge}_{15}\text{Sb}_{85}$ and $\text{Ge}_{15}\text{Te}_{85}$ near their glass transition temperatures. Both systems are influenced by x-ray irradiation, yet their responses differ markedly despite their similar composition. $\text{Ge}_{15}\text{Sb}_{85}$ undergoes a rapid transition to a stationary yielding regime within the exposure time, characterized by a KWW parameter $\beta \lesssim 1$. By contrast, $\text{Ge}_{15}\text{Te}_{85}$ exhibits strong dose-dependent evolution, including aging and a progressive shift of the shape parameter β from compressed ($\beta > 1$) to stretched ($\beta < 1$). This transition is consistent with the emergence of liquidlike collective motion, as supported by de Gennes narrowing observed in the wave vector q -dependence of the dynamics. This glass does not reach a stationary regime within experimental timescales, indicating that local yielding requires thousands of seconds. This resistance to photoinduced yielding in $\text{Ge}_{15}\text{Te}_{85}$ is consistent with its heavier atomic constituents, which enhance structural rigidity and demand much higher deposited doses to induce local transformations. These findings align with its higher amorphous thermal stability and slow crystallization kinetics [28], in contrast to $\text{Ge}_{15}\text{Sb}_{85}$ which exhibits rapid crystallization kinetics and less covalent bonding [22]. Overall, neither chemical composition alone nor bonding character alone dictates the x-ray-induced response of the non-PCM $\text{Ge}_{15}\text{Te}_{85}$ and PCM $\text{Ge}_{15}\text{Sb}_{85}$; instead, it emerges from the interplay between irradiation effects and intrinsic relaxation pathways. Finally, we note that such beam-induced dynamics may provide a novel means of probing β relaxations in PCMs below T_g . Previous studies have shown that PCMs such as $\text{Ge}_2\text{Sb}_2\text{Te}_5$, GeTe , and AIST exhibit pronounced excess wings in the loss modulus associated with β relaxations, in stark contrast to those chalcogenide non-PCMs whose excess wings are vanishingly small [48].

Beyond the specific phenomenology reported here, our results highlight the unique capability of XPCS to access

direct measurements of atomic and nanoscale dynamics under photoexcitation. In contrast to previous ultrafast diffraction and spectroscopic studies in phase-change materials, which capture averaged structural responses [33–36], XPCS directly probes the temporal decorrelation of the speckle pattern, providing quantitative insight into the microscopic relaxation pathways and yielding dynamics. The advent of next-generation light sources and detector technologies—such as high-repetition-rate XFELs, diffraction-limited storage rings, and fast area detectors—will further extend this approach, enabling pump–probe XPCS experiments with both femtosecond time resolution and statistical sensitivity to subtle decorrelation processes. These advances will open the way to directly resolve photoinduced atomic-scale dynamics in phase-change and related functional materials, bridging the gap between averaged ultrafast structural probes and truly correlation-resolved measurements.

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G.B. and B.R. conceived and designed the project. G.B., A.C., I.F., T.F., J.M., A.R., B.R., M.S., S.W., and F.W. performed the experiments. J.M., R.E., and A.P. carried out sample fabrication. I.F. analyzed the data. G.B., B.R., and I.F. wrote the manuscript with contributions from all the authors. All the authors contributed to the discussion of the results, reviewed the data, and provided feedback on the manuscript.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

- [1] J. Baruchel, J.-L. Hodeau, and M. S. Lehmann, *Neutron and Synchrotron Radiation for Condensed Matter Studies, Theory, Instruments and Methods* (Springer, 1993), Vol. 1.
- [2] A. Madsen, A. Flueraşu, and B. Ruta, Structural dynamics of materials probed by X-ray photon correlation spectroscopy, in *Synchrotron Light Sources and Free-Electron Lasers: Accelerator Physics, Instrumentation and Science Applications*, edited by E. Jaeschke, S. Khan, J. R. Schneider, and J. B. Hastings (Springer International Publishing, 2016), pp. 1617–1641.
- [3] M. Leitner, B. Sepiol, L.-M. Stadler, B. Pfau, and G. Vogl, Atomic diffusion studied with coherent X-rays, *Nat. Mater.* **8**, 717 (2009).
- [4] B. Ruta, Y. Chushkin, G. Monaco, L. Cipelletti, E. Pineda, P. Bruna, V. M. Giordano, and M. Gonzalez-Silveira, Atomic-scale relaxation dynamics and aging in a metallic glass probed by X-ray photon correlation spectroscopy, *Phys. Rev. Lett.* **109**, 165701 (2012).
- [5] J. Shen, F. Yang, A. Cornet, E. Pineda, K. Martens, Y. Chushkin, F. Zontone, I. Festi, A. Ronca, N. Neuber, *et al.*, Length-scale dependence of the anomalous atomic motion in metallic glasses, *Nat. Phys.* **22**, 672 (2026).
- [6] B. Ruta, S. Hechler, N. Neuber, D. Orsi, L. Cristofolini, O. Gross, B. Bochtler, M. Frey, A. Kuball, S. S. Riegler, M. Stolpe, Z. Evenson, C. Gutt, F. Westermeier, R. Busch, and I. Gallino,

- Wave-vector dependence of the dynamics in supercooled metallic liquids, *Phys. Rev. Lett.* **125**, 055701 (2020).
- [7] A. Das, P. M. Derlet, C. Liu, E. M. Dufresne, and R. Maaß, Stress breaks universal aging behavior in a metallic glass, *Nat. Commun.* **10**, 5006 (2019).
- [8] M. Frey, N. Neuber, S. S. Riegler, A. Cornet, Y. Chushkin, F. Zontone, L. M. Ruschel, B. Adam, M. Nabahat, F. Yang, *et al.*, Liquid-like versus stress-driven dynamics in a metallic glass former observed by temperature scanning X-ray photon correlation spectroscopy, *Nat. Commun.* **16**, 4429 (2025).
- [9] N. Amini, F. Yang, E. Pineda, B. Ruta, M. Sprung, and A. Meyer, Intrinsic relaxation in a supercooled ZrTiNiCuBe glass forming liquid, *Phys. Rev. Mater.* **5**, 055601 (2021).
- [10] B. Ruta, F. Zontone, Y. Chushkin, G. Baldi, G. Pintori, G. Monaco, B. Rufflé, and W. Kob, Hard X-rays as pump and probe of atomic motion in oxide glasses, *Sci. Rep.* **7**, 3962 (2017).
- [11] G. Pintori, G. Baldi, B. Ruta, and G. Monaco, Relaxation dynamics induced in glasses by absorption of hard X-ray photons, *Phys. Rev. B* **99**, 224206 (2019).
- [12] F. Dallari, G. Pintori, G. Baldi, A. Martinelli, B. Ruta, M. Sprung, and G. Monaco, X-rays induced atomic dynamics in a lithium-borate glass, *Condens. Matter Phys.* **22**, 43606 (2019).
- [13] K. Holzweber, C. Tietz, T. M. Fritz, B. Sepiol, and M. Leitner, Beam-induced atomic motion in alkali borate glasses, *Phys. Rev. B* **100**, 214305 (2019).
- [14] J. Li, M. Madhavi, S. Jeppson, L. Zhong, E. M. Dufresne, B. Aitken, S. Sen, and R. Kukreja, Observation of collective molecular dynamics in a chalcogenide glass: Results from X-ray photon correlation spectroscopy, *J. Phys. Chem. B* **126**, 5320 (2022).
- [15] F. Dallari, A. Martinelli, F. Caporaletti, M. Sprung, G. Baldi, and G. Monaco, Stochastic atomic acceleration during the X-ray-induced fluidization of a silica glass, *Proc. Natl. Acad. Sci. USA* **120**, e2213182120 (2023).
- [16] E. Alfinelli, F. Caporaletti, F. Dallari, A. Martinelli, G. Monaco, B. Ruta, M. Sprung, M. Zanatta, and G. Baldi, Amorphous-amorphous transformation induced in glasses by intense X-ray beams, *Phys. Rev. B* **107**, 054202 (2023).
- [17] J. Baglioni, A. Martinelli, P. Sun, F. Dallari, F. Westermeier, M. Sprung, G. Grübel, and G. Monaco, Uniqueness of glasses prepared via X-ray induced yielding, *Rep. Prog. Phys.* **87**, 120503 (2024).
- [18] A. Martinelli, F. Caporaletti, F. Dallari, M. Sprung, F. Westermeier, G. Baldi, and G. Monaco, Reaching the yield point of a glass during X-ray irradiation, *Phys. Rev. X* **13**, 041031 (2023).
- [19] S. Wei, P. Lucas, and C. Angell, Phase-change materials: The view from the liquid phase and the metallicity parameter, *MRS Bull.* **44**, 691 (2019).
- [20] A. Piarristeguy, P. Noé, and F. Hippert, Verres de chalcogénures pour le stockage de l'information, *Reflets phys.* **74**, 58 (2022).
- [21] Y. Cheng, Q. Yang, J. Wang, T. Dimitriadis, M. Schumacher, H. Zhang, M. J. Müller, N. Amini, F. Yang, A. Schoekel, J. Pries, R. Mazzarello, M. Wuttig, H.-B. Yu, and S. Wei, Highly tunable β -relaxation enables the tailoring of crystallization in phase-change materials, *Nat. Commun.* **13**, 7352 (2022).
- [22] J. Moesgaard, T. Fujita, and S. Wei, Unveiling the boson peaks in amorphous phase-change materials, *J. Phys.: Condens. Matter* **37**, 025101 (2025).
- [23] J. Y. Raty, W. Zhang, J. Luckas, C. Chen, R. Mazzarello, C. Bichara, and M. Wuttig, Aging mechanisms in amorphous phase-change materials, *Nat. Commun.* **6**, 7467 (2015).
- [24] M. Chen, K. A. Rubin, and R. W. Barton, Compound materials for reversible, phase-change optical data storage, *Appl. Phys. Lett.* **49**, 502 (1986).
- [25] S. Wei, P. Lucas, and C. Angell, Phase change alloy viscosities down to T_g using Adam-Gibbs-equation fittings to excess entropy data: A fragile-to-strong transition, *J. Appl. Phys.* **118**, 034903 (2015).
- [26] H. Weber, J. Orava, I. Kaban, J. Pries, and A. L. Greer, Correlating ultrafast calorimetry, viscosity, and structural measurements in liquid GeTe and Ge₁₅Te₈₅, *Phys. Rev. Mater.* **2**, 093405 (2018).
- [27] S. Wei, M. Stolpe, O. Gross, W. Hembree, S. Hechler, J. Bednarcik, R. Busch, and P. Lucas, Structural evolution on medium-range-order during the fragile-strong transition in Ge₁₅Te₈₅, *Acta Mater.* **129**, 259 (2017).
- [28] R. Sengottaiyan, N. Saxena, K. D. Shukla, and A. Manivannan, Temperature dependent structural evolution and crystallization properties of thin Ge₁₅Te₈₅ film revealed by *in situ* resistance, X-ray diffraction and scanning electron microscopic studies, *J. Phys. D: Appl. Phys.* **53**, 025108 (2020).
- [29] P. Lucas, S. Wei, and C. Angell, Liquid-liquid phase transitions in glass-forming systems and their implications for memory technology, *Int. J. Appl. Glass Sci.* **11**, 236 (2020).
- [30] J. Pries, S. Wei, F. Hoff, P. Lucas, and M. Wuttig, Control of effective cooling rate upon magnetron sputter deposition of glassy Ge₁₅Te₈₅, *Scr. Mater.* **178**, 223 (2020).
- [31] J. Pries, Y. Yu, P. Kerres, M. Häser, S. Steinberg, F. Gladisch, S. Wei, P. Lucas, and M. Wuttig, Approaching the glass transition temperature of GeTe by crystallizing Ge₁₅Te₈₅, *Phys. Status Solidi RRL* **15**, 2000478 (2021).
- [32] T. Fujita, Y. Chen, Y. Kono, S. Takahashi, H. Kasai, D. Campi, M. Bernasconi, K. Ohara, H. Yumoto, T. Koyama, H. Yamazaki, Y. Senba, H. Ohashi, I. Inoue, Y. Hayashi, M. Yabashi, E. Nishibori, R. Mazzarello, and S. Wei, Pressure-induced reversal of Peierls-like distortions elicits the polyamorphic transition in GeTe and GeSe, *Nat. Commun.* **14**, 7851 (2023).
- [33] L. Waldecker, T. A. Miller, M. Rudé, R. Bertoni, J. Osmond, V. Pruneri, R. E. Simpson, R. Ernstorfer, and S. Wall, Time-domain separation of optical properties from structural transitions in resonantly bonded materials, *Nat. Mater.* **14**, 991 (2015).
- [34] E. Matsubara, S. Okada, T. Ichitsubo, T. Kawaguchi, A. Hirata, P. F. Guan, K. Tokuda, K. Tanimura, T. Matsunaga, M. W. Chen, and N. Yamada, Initial atomic motion immediately following femtosecond-laser excitation in phase-change materials, *Phys. Rev. Lett.* **117**, 135501 (2016).
- [35] P. Zalden, F. Quirin, M. Schumacher, J. Siegel, S. Wei, A. Koc, M. Nicoul, M. Trigo, P. Andreasson, H. Enquist, M. J. Shu, T. Pardini, M. Chollet, D. Zhu, H. Lemke, I. Ronneberger, J. Larsson, A. M. Lindenberg, H. E. Fischer, S. Hau-Riege, *et al.*, Femtosecond x-ray diffraction reveals a liquid-liquid phase transition in phase-change materials, *Science* **364**, 1062 (2019).
- [36] Y. Qi, N. Chen, T. Vasileiadis, D. Zahn, H. Seiler, X. Li, and R. Ernstorfer, Photoinduced ultrafast transition of the local

- correlated structure in chalcogenide phase-change materials, *Phys. Rev. Lett.* **129**, 135701 (2022).
- [37] G. Grübel and F. Zontone, Correlation spectroscopy with coherent X-rays, *J. Alloys Compd.* **362**, 3 (2004), Proceedings of the Sixth International School and Symposium on Synchrotron Radiation in Natural Science (ISSRNS).
- [38] J. C. Phillips, Stretched exponential relaxation in molecular and electronic glasses, *Rep. Prog. Phys.* **59**, 1133 (1996).
- [39] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/d39x-d2hv> for details on absorbed-dose calculations, data treatment, and additional results on beam-induced dynamics.
- [40] Z. Evenson, B. Ruta, S. Hechler, M. Stolpe, E. Pineda, I. Gallino, and R. Busch, X-ray photon correlation spectroscopy reveals intermittent aging dynamics in a metallic glass, *Phys. Rev. Lett.* **115**, 175701 (2015).
- [41] P. De Gennes, Liquid dynamics and inelastic scattering of neutrons, *Physica* **25**, 825 (1959).
- [42] N. Neuber, O. Gross, M. Frey, B. Bochtler, A. Kuball, S. Hechler, F. Yang, E. Pineda, F. Westermeier, M. Sprung, F. Schäfer, I. Gallino, R. Busch, and B. Ruta, Disentangling structural and kinetic components of the α -relaxation in supercooled metallic liquids, *Commun. Phys.* **5**, 316 (2022).
- [43] V. Novikov, K. Schweizer, and A. Sokolov, Coherent neutron scattering and collective dynamics on mesoscale, *J. Chem. Phys.* **138**, 164508 (2013).
- [44] B. Ruta, G. Baldi, Y. Chushkin, B. Rufflé, L. Cristofolini, A. Fontana, M. Zanatta, and F. Nazzani, Revealing the fast atomic motion of network glasses, *Nat. Commun.* **5**, 3939 (2014).
- [45] R. Shekhawat, H. Pamuluri, V. Erkkara Madhavan, and K. Ramesh, Structural transformation and phase change properties of Se substituted GeTe, *Sci. Rep.* **11**, 7604 (2021).
- [46] E. Martin and C. Massobrio, Amorphous structures and statistical sampling in first-principles molecular dynamics: The prototypical case of glassy GeSe₃, *J. Non-Cryst. Solids* **653**, 123415 (2025).
- [47] J. Kalikka, J. Akola, R. O. Jones, S. Kohara, and T. Usuki, Amorphous Ge₁₅Te₈₅: Density functional, high-energy X-ray and neutron diffraction study, *J. Phys.: Condens. Matter* **24**, 015802 (2012).
- [48] S.-X. Peng, Y. Cheng, J. Pries, S. Wei, H.-B. Yu, and M. Wuttig, Uncovering β -relaxations in amorphous phase-change materials, *Sci. Adv.* **6**, eaay6726 (2020).